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SUPPLEMENTARY INFORMATION

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Substantial global carbon uptake by cement carbonation

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Supplementary Methods

1 *Cement chemistry*

1.1 Cement constituents and hydration reactions

The primary constituents of cement are calcium oxide (CaO), tricalcium silicate (C_3S), dicalcium silicate (C_2S), tricalcium aluminate (C_3A), tetracalcium aluminoferrite (C_4AF), gypsum ($C\underline{S}H_2$), as well as other, lesser constituents such as alkali, MgO, Na_2O, Ka_2O, heavy metal, etc.[1,2] as well as additions of limestone, fly-ash, silica-fume, and Ground Granulated Blast-Furnace slag (GGBF-slag)depending on proprietary recipes and use[3]. Following conventions in the cement, we abbreviate these ingredients: CaO = C; SiO_2 = S; Al_2O_3 = A; Fe_2O_3 = F; H_2O = H; SO_3 = $\underline{S}$. Cement hydration reactions form calcium hydroxide, calcium silicate hydrates, and other hydration products as follows[4]:

$$CaO + H_2O \rightarrow Ca\left(OH\right)_2$$

$$2C_3S + 6H \rightarrow C_3\underline{S}_2H_3 + 3Ca(OH)_2$$

$$C_2S + 4H \rightarrow C_3\underline{S}_2H_3 + Ca(OH)_2$$

$$C_3A + 3C\underline{S}H_2 + 26H \rightarrow C_6AS_3H_{32}$$

$$2C_3A + 3C_6A\underline{S}_3H_{32} + 22H \rightarrow 3C_4ASH_{18}$$

$$C_4AF + 3C\underline{S}H_2 + 3H \rightarrow 6(A,F)\underline{S}_3 + (A,F)H_3 + Ca(OH)_2$$

Upon wetting of cement materials such as mortar and concrete, the main constituents are hydrated by the above reactions to produce CH [$Ca(OH)_2$ or unhydrated CaO•H_2O], CSH [2(CaO)•SiO_2•0.9-1.25(H_2O), and/or; CaO•SiO_2•1.1(H_2O), and/or; 0.8-1.5(CaO)•SiO_2•1.0-2.5(H_2O)], CAH [more complex than C-S-H], AF_t ($C_3AS_3H_{30-32}$), AFm (C_2ASH_{12}), C_3AH_6 (3CaO•Al_2O_3•6 H_2O), MgO•SiO_2•x(H_2O), and so on[1]. In turn, these alkaline hydrated products are unstable and progressively react with the carbon dioxide in the air[5].

1.2 Carbon uptake by cement carbonation

Carbonation is a complicated physicochemical reaction between CO_2 and hydrated cement products in the presence of pore water, which ultimately sequesters carbon in cement[6,7,8]. Eventually, the carbonation process weakens structures and materials, as the carbonation reactions dissolve cement from concrete and mortar and return it to the constituent ingredients of calcium carbonate, and hydrated silica, alumina, and iron oxide[9]. The main chemical reactions of cement carbonation are showed in method.

2 Cement production and consumption

Cement production data for China, the U.S., Europe, and rest of world from 1930 to 2013 is from the US Geological Survey[10] *(Supplementary Data 1)*. Concrete and mortar account for most of the produced cement (roughly 70% and 30%, respectively; *Supplementary Data 2*), with a small fraction related to cement kiln dust (CKD) generated during clinker production.

On average, 69.7%-86.0% of cement is used for concrete, with little variation among world regions (*Supplementary Data 2*). Based on survey statistics of 1144 samples of civil engineering projects between 1980 and2012, we estimate that 69.7% of cement produced in China is used for concrete, 28.8% for mortar, and 1.5% loss in construction waste. In the U.S., sources suggest that 86.0% of cement is used for concrete[11]. In Europe, the proportion falls in between China and the U.S.: we estimate that 71.1% of European cement is used for concrete based on European Ready Mixed Concrete Organization (ERMCO) statistics[12] and research in Nordic countries in 2003[13] *(Supplementary Table 1)*. Owing to a lack of data for other regions, we assume the central estimate of cement used for concrete (i.e. Europe's) applies.

Of the produced concrete and mortar, roughly 1-3% is wasted during construction, but the shelf life of cement is only 3 to 6 months on average, such that the actual cement consumed in a given year is 97%-99% of the total cement produced *(Supplementary Table 1)*. Prior studies show only minor variation in this utilization fraction across regions (<4.5%; *Supplementary Data3)*. Cement kiln dust (CKD) related to clinker production[14,15] (*Supplementary Table 4*) will absorb carbon dioxide during landfill/waste treatment[16,17], as will cement waste generated in construction.

3 Process model of cement carbonation

Using production data as described in section 2, we adopt a life cycle assessment (LCA) method to estimate carbon up take by cement materials over time *(Supplementary Figure1).* Total carbon uptake of cement is calculated by adding carbon uptake by concrete cement, carbon uptake by mortar cement, carbon uptake by construction cement waste, and carbon uptake by cement kiln dust (see method).

3.1 Carbon uptake by concrete cement

We divide the concrete life cycle into three phases: service life (e.g., in buildings), demolition, and secondary use (including both disposal in a landfill and recycling)[18]. In case, we calculate CO_2 uptake of concrete by adding carbon uptake during the service life, carbon uptake during the demolition and carbon uptake during the secondary use stage.

3.1.1 Service life

Concrete categories

We further break down cement utilization for different categories of concrete[19]. For China, our estimates are based on the average cement consumption from 1999 to 2002 in *China Statistical Yearbook on Construction*, with concrete used for residential construction also incorporating data on proportion of housing floor types completed areas from 1996 to 2012 published in the *China Statistical Yearbook on Construction*. The concrete utilization category in THE U.S. is based on statistics of apparent cement consumption in the United States from 1975 to 2013[10] and a report by the Portland Cement Association[9]. Categories of concrete used in Europe are based on ERMCO Statistics 2001-2013[12] and the studies of Pade and Guimaraes[13,18]. Again, we assume the rest of world uses concrete as Europe does (*Supplementary Data*5).

Concrete strength classes

We estimate strength classes of Chinese concretes based on the survey statistics for U.S. concrete based on data from ERMCO Statistics 2001-2013[12] and studies by Low and Nisbet[20,21], and for European and rest of world concretes based on ERMCO Statistics 2001-2013[12] and the study of Pade and Guimaraes in Nordic countries[18] (*Supplementary Table 2 and Supplementary Data* 6).

Concrete cement content

The cement content for concrete is the mass of cement used in one cubic meter of concrete (kg/m^3) (*Supplementary Data* 7). We use Chinese cement contents from the *Construction and Installation Engineering Budget Manual* and *Concrete Mix Proportion Quick Manual*[22,23], U.S. cement contents from the same ERMCO Statistics 2001-2013 as well as a study by Low[20], and European and rest of world cement contents from EN 206-1:2000 and averages in ERMCO Statistics 2001-2013[12,13,24],

Exposure conditions, CO_2 concentrations, and additives.

Following the study of Pade and Guimareas, we estimate carbon uptake of cement materials' carbonation under five different categories of exposure conditions: exposed, sheltered, indoors, wet, and buried[18]. Specifically, relative humidity, ambient CO_2 concentration[35,36], and additives have been shown to affect carbonation rate coefficients[25]. The range of applicable conditions are estimated based on the previously referenced, region-specific studies and survey statistics[9,18]. Because the concrete carbonation depth is proportional to $\sqrt{CO_2\ concentration}$ [25,26], we apply the correction factors shown in *Supplementary Table 3*. Similarly, additives to cement and concrete may affect the carbonation rate[18], and we take this into account by applying other correction factors as shown in *Supplementary Table 4*.

Coating and coverings

Studies have shown that application of surface coating and coverings such as paints can reduce the rates of cement carbonation by 10-30%[27,19]. However, some studies have also shown that paint does not substantially reduce the carbonation depth[28,29]. Covering concrete with mortar has also been shown to reduce the concrete carbonation rate, and previous studies have applied the carbonation rate correction coefficient to calculate carbonation depth of concrete[8,27,30]. Recent study showed that paint coating can reduce 28-day carbonation depth of concrete by 46%[31]. The accelerated carbonation test of prime impermeable anti-deteriorating coating in Republic of Korea for 7days, 14days, 28 days, and 56 days showed that the carbonation correction coefficients of coating ranged from 24% to 42%[32].

The paints and other coatings may protect against carbonation for 1-2 years, and the protection will diminish over time if these coverings are not reapplied[33]. However, there are no long-term studies of the extent to which carbonization is delayed over multiple years. Given this uncertainty, we assess carbonation using carbonation correction coefficients meant to reflect the potential effects of coatings, including decreases in carbonation rates of up to 50% over the life cycle of concretes *(Supplementary Table 5)*.

Concrete carbonation rates

Using our estimates of concrete category, cement content, exposure conditions, additives and coatings, we use relevant concrete carbonation rate coefficients from various region-specific references[18,9] *(Supplementary Table 6 and Supplementary Data 8)*. We further revised concrete carbonation rate coefficients in China and other countries considering the impacts of *compressive* *strength class and exposure conditions ($\beta_{c\sec}$)[34], cement addition (β_{ad})[19], CO_2 concentration (β_{CO_2})[25,26], and coating and cover (β_{CC})[29,35].*

Service life duration

Based on the previous, region-specific references, we estimate concrete service life *(t_l)* in China is 35 years (ranging from 4-73 years), the duration of the demolition stage *(t_d)* is 0.4 years (ranging from 0.1-0.8 years), and the duration of the secondary use stage *(t_s)* is 64.6 years[36,37]. In Europe, service life is estimated to be 70 years (ranging from 50-90 years), demolition stage is 0.4 years (ranging from 0.1-0.7 years), and secondary use stage is 29.6 years[18,24]. In the U.S., service life is estimated to be 65 years (ranging from 56-84 years), the demolition stage is again 0.4 (0.1-0.7) years, and the secondary use stage is 34.6 years[38]. In the rest of world region, we estimate the duration of concrete service life is 40 (10-90) years, demolition stage is 0.4 (0.1-1.0) years, and secondary use stage is 59.6 years[39] *(Supplementary Table 7, Supplementary Data9 and Supplementary Data 10).*

Carbonation depth

We use the applicable carbonation rate coefficients and exposure times to calculate the carbonation depth of concrete in each strength class and set of exposure conditions using Fick's diffusion law[18].

Exposed surface area

We estimate the exposed surface area of concrete in the U.S., China, Europe, and rest of world based on average typical thickness of concrete structures in the literature[9,18,24] *(Supplementary Data 11)*.

Volume of carbonated concrete and carbonated cement

We calculate the carbonated concrete volume by exposed surface area and carbonation depth. The carbonated cement in service life can then be calculated by the cement content of concrete in different strength classes (kg cement/m^3), clinker to cement ratio, average CaO content of clinker in cement[40], the proportion of CaO within fully carbonated cement that converts to $CaCO_3$[9,18,34,41-43], and the ratio of C element to CaO (see method).

Annual carbon uptake by concrete in service

Finally, we combine the results of the above calculations to calculate the annual carbon uptake in year t_l as the cumulative carbon uptake in year t_l minus the cumulative carbon uptake in year t_l-1.

3.1.2 Demolition stage

During demolition stage, the size of waste concrete pieces is determined according to the intended secondary use (e.g., disposal in a landfill, stacking, or recycling). If the waste concrete will be diverted to a landfill or dump, it will be transported to the landfill and dump sites after demolition, where it may be further crushed for steel recycling, and probably stockpiled for a short time period. Even so, some relatively large pieces of concrete (e.g., with diameters of 0.5 m) may be buried in a landfill. If the concrete is recycled as new cement or asphalt concrete aggregates, road base, backfills (e.g., in highway embankments), pieces will be transported to recycling plants and crushed into different particle sizes according to the intended secondary use *(Supplementary Figure 2)*. The surface area of exposed concrete, duration of exposure, and exposure conditions of demolished concretes will therefore vary substantially. The fate of demolition waste in different regions is taken from different literature sources[18,24,38,39,44-46] *(Supplementary Table8)*.

Size and surface area of concrete pieces

The range and particle size distribution of different types of demolished concrete in each region is estimated in *Supplementary Table9*. In China, surveys of 179 demolition projects in 35 large cities were summarized. According to the China National and Industrial Standard, the maximum particle size of concrete recycled into new cement, asphalt concrete aggregates, or highway base is 32.5 mm, and the maximum particle size recycled for second-level road aggregates is 53 mm[47]. We use published particle size distributions for Nordic countries to estimate distributions in Europe[18,48], and a combination of European and Chinese distribution to estimate values applicable to the U.S.. Finally, we adopt particle size distributions in Japan and South Korea in estimating carbon uptake for demolished concretes in the rest of world [39,46]*(Supplementary Data 12)*.

Exposure time

The average exposure time of concrete during the demolition stage is estimated about 0.4 years in whole world *(Supplementary Table7)*. In China, this average exposure time is derived from our field survey data *(Supplementary Data 10)*, which showed 1-4 weeks for building demolition and crushing processes related to steel recycling, 1-24 weeks for stockpiling before transporting to landfill sites or recycling plants, and an additional 4-16 weeks for recycled concrete products stockpiled before secondary use. According to literature sources, the durations are similar in Japan, Korea, and Europe[34,46,48].

Carbonation of demolished concrete

The carbonation fraction of demolished concrete is calculated according to particle size distributions and carbonation depths using the Fick's diffusion law[24,49]. Using the fraction of concrete that will undergo carbonation, we next calculate the mass of concrete cement carbonated during the demolition stage by the concrete cement carbonated during demolition for each concrete strength class *i*, the cement consumed for each strength class *i* of concrete, the concrete cement carbonated during service life for each strength class *i*, the fraction of carbonated cement in concrete strength class *i* in the demolition stage, and the total mass of concrete cement carbonated in the demolition stage (see method).

Total carbon uptake during demolition stage

Finally, we estimate total carbon uptake during the demolition stage based on cement carbonated in demolition stage and carbonation fraction of differently treated concretes[18,34,42].

3.1.3 Secondary use stage

It is estimated that more than 91% of crushed concrete particles worldwide are buried, either in landfills or as part of their recycled use such as for road base or backfill aggregates *(Supplementary Table 9).* The proportion diverted to landfills and dumps tends to be higher instill-developing countries like China. Burial ensures that little of this concrete is exposed to air[42], and concrete recycled as aggregate for new cement or asphalt concrete are bound in concrete (making it difficult to evaluate its susceptibility to continued carbonation). Thus, we assume that concretes diverted to landfills or these secondary uses cannot be further carbonated.

Carbonation depth in secondary use stages

The total carbonation depth in demolition stage and secondary use stage can be estimated by carbonated depth in demolition stage plus new carbonation depth during the secondary use stage *(Supplementary Figure 3a and Supplementary Figure 3b)*. There is the time lag (Δt_i) for the same carbonation depth from air exposure condition to buried condition *(Supplementary Figure 3c)* using the Fick's diffusion law.

Fraction carbonized

The estimations details are showed in method.

Cumulative and annual carbon uptake during the secondary use stage

The estimations details are showed in method.

3.2 Carbon uptake by mortar cement

3.2.1 Cement utilization for mortars

The cement utilization for mortars is showed in *Supplementary Table 10* [50]. Most mortar is used for rendering, plastering and decorating (e.g., covering the exterior surfaces of concrete structures and walls)[51,52]. The proportion of mortar cement for repairing in different building types to total cement consumption ranges from 0.21% to 3.37%[9]. Our Chinese survey data *(Supplementary Data13)* indicates that about 70% of mortar cement used for rendering, plastering and decorating, 18% of mortar cement used for masonry, 12% for maintenance and repairing, and very small quantities for all other uses. In the U.S., USGS end-use statistics show similar proportions of cement used for different types of mortar[11]. Because we do not have other region-specific data, we assume Chinese utilization rates apply Europe and rest of world *(Supplementary Data14)*.

3.2.2 The typical thickness of cement mortar utilization

The typical thickness of cement mortar utilization is showed in *(Supplementary Table 10)*[27,50-52].

3.2.3 Carbonation rate coefficients of cement mortar

Cement mortars have been shown to undergo carbonation at a faster rate than concrete because they have a lower cement content, higher water/cement ratios, and aggregates of finer grain size (maximum grain size of 2–4 mm)[53,54]. However, there are relatively few studies of mortar carbonation rates and depth. Because mortar is essentially concrete with fine-grain aggregate, the carbonation principles of mortar should be similar to those of low strength class concrete (<C15), which on average have carbonation rate coefficients of between 6.1 mm/$\sqrt{\text{year}}$ and 36.8 mm/$\sqrt{\text{year}}$ in outdoor and indoor exposure conditions, respectively (in temperate climate conditions and according to our field survey and experiment data using the 1% alcohol phenolphthalein solution; see Supplementary Data15). Carbonation depth will increase if the cement contains more additives[30].

3.2.4 Carbon uptake by mortar cements

The large exposure area and thin layers of mortar cement translates into rapid carbonation. We calculate annual carbon uptake based on the proportion of annual carbonation depth[55,56], and estimate carbon uptake as the sum of uptake by adding the carbon uptake of rendering and plastering mortar, carbon uptake of masonry mortar, and carbon uptake of maintain and repairing mortar (see method). The proportion of *CaO* within fully carbonated mortar cement that converts to $CaCO_3$ is showed in *Supplementary Data 16*. The survey data for masonry walls are covered by rendering mortar on both sides, only inside, and no rendering cover is showed in *Supplementary Data 17*. The carbon uptake calculation methods for both rendering and plastering mortar and maintain and repairing mortar are showed in method.

The carbon uptake by masonry cement mortar can is calculated as

$$C_{r\,mat} = C_{mbt} + C_{mot} + C_{mnt} \qquad \textit{[Supplementary eq.1]}$$

where C_{mbt} is carbon uptake by masonry mortar of walls with both sides rendered, C_{mot} is carbon uptake by masonry mortar of walls with one side rendered, and C_{mnt} is carbon uptake by masonry mortar of walls with no rendering. The carbon uptake calculation method of C_{mbt}, C_{mot}, and C_{mnt} is similar as that of rendering and plastering mortar by considering wall thickness and demolition effects.

We calculate carbonation of masonry mortar for walls with both sides rendered (C_{mbt}) by

$$d_{mb} = \begin{cases} 0 & (t \le t_r) \\ 2\left(K_m \times \sqrt{t} - d_{Trp}\right) & (t \ge t_r) \end{cases} \qquad \textit{[Supplementary eq.2]}$$

$$f_{mbt} = \begin{cases} 0 & (t \le t_r) \\ \left(d_{mbt} - d_{mb(t-1)}\right) / d_w \times 100\% & (t_r \le t \le t_{sl}) \\ 100\% - \left(d_{mbt_l} - 2d_{Trp}\right) / d_w \times 100\% & (t = t_{sl} + 1) \end{cases} \qquad \textit{[Supplementary eq.3]}$$

$$C_{mbt} = W_m \times r_{rm} \times r_b \times f_{mbt} \times C_{clinker} \times f_{CaO} \times \gamma_1 \times M_r \qquad \textit{[Supplementary eq.4]}$$

where d_{mb} is the total carbonation depth of masonry mortar of wall with both sides rendered, K_m is the carbonation rate coefficient of mortar, *t* is the exposure time of masonry mortar after construction, and t_{r} is the time of full carbonation of render mortar in d_{Trp} thickness, d_{Trp} is the thickness of render mortar on masonry wall, f_{mbt} is the annual carbonation percent of cement used for masonry mortar with both sides rendered on year t, d_{mbt} and $d_{mb(t-1)}$ are carbonation depth of masonry mortar with both sides rendered at t and t-1 times, respectively; d_w is the thickness of masonry wall, d_{mbt_l} is the carbonation depth of a masonry mortar with both sides render in service life years (t_l), C_{mbt} is the annual carbon uptake of cement for masonry mortar with both sides render in year t, W_m is the cement for mortar, and r_{rm} is the percentage of masonry mortar cement in total mortar cement.

We then calculate carbonation of masonry mortar for walls with one side rendered (C_{mot}) by

$$d_{mo} = \begin{cases} K_m \times \sqrt{t} & (t \le t_r) \\ K_m \times \sqrt{t} + \left(K_m \times \sqrt{t} - d_{Trp}\right) & (t_{\mathrm{r}} \le t \le t_{sl}) \end{cases} \quad \textit{[Supplementary eq.5]}$$

$$f_{mot} = \begin{cases} \left(d_{mot} - d_{mo(t-1)}\right) / \mathrm{d_w} \times 100\% & (t_{\mathrm{r}} \le t \le t_{sl}) \\ 100\% - \left(2d_{mot_l} - d_{Trp}\right) / \mathrm{d_w} \times 100\% & (t = t_{sl} + 1) \end{cases} \quad \textit{[Supplementary eq.6]}$$

$$C_{mot} = W_m \times r_{rm} \times r_o \times f_{mot} \times C_{clinker} \times f_{CaO} \times \gamma_1 \times M_r \quad \textit{[Supplementary eq.7]}$$

where d_{mo} is total carbonation depth of masonry mortar of wall with one side rendered, t_{r} is the full carbonation time of render mortar outside of the masonry wall, f_{mot} is the annual carbonation percent of cement used for masonry mortar with one side rendered, d_{mot} and $d_{mo(t-1)}$ are total carbonation depth of masonry mortar with one side rendered at t and t-1 times, respectively; d_{mot_l} is the carbonation depth of masonry mortar with one side rendered during the service life years (t_l), d_{Trp} is the thickness of render mortar of the masonry wall, C_{mot} is the annual carbon uptake of cement for masonry mortar with one side render on year t.

Finally, we calculate carbonation of masonry mortar without rendering (C_{mnt}) by

$$d_{mn} = 2K_m \times \sqrt{t} \quad \textit{[Supplementary eq. 8]}$$

$$f_{mnt} = \begin{cases} 2\left(d_{mnt} - d_{mn(t-1)}\right) / \mathrm{d_w} \times 100\% & (t \le t_{sl}) \\ 100\% - 2d_{mnt_l} / \mathrm{d_w} \times 100\% & (t = t_{sl} + 1) \end{cases} \quad \textit{[Supplementary eq.9]}$$

$$C_{mnt} = W_m \times r_{rm} \times r_n \times f_{mnt} \times C_{clinker} \times f_{CaO} \times \gamma_1 \times M_r \quad \textit{[Supplementary eq.10]}$$

where d_{mn} is the total carbonation depth of masonry mortar of wall without wall render, f_{mnt} is the annual carbonation percent of cement used for masonry mortar without wall render in year t. d_{mnt} and $d_{mn(t-1)}$ are carbonation depth of masonry mortar with no wall render at t and t-1 times, respectively; d_{mnt_l} is the total carbonation depth of masonry mortar with no wall render during the service life years (t_l), C_{mnt} is the annual carbon uptake of cement for masonry mortar with no wall render in year t.

3.3 Carbon uptake by cement in construction wastes

Cement wasted during construction accounts for 1% to 3% (average 1.5%) of total cement consumption according to construction budget standards[22] and survey data[58]. Most of this waste is in small pieces and will be recycled as backfill or landfilled after the completion of building projects. Of these wastes, about 45% is concrete and 55% is mortar[37,59]. Given the small sizes of pieces, the waste mortar is assumed to completely carbonate in the first year, and concrete wastes are assumed to completely carbonate over the following 5 years (ranging from 1 to 10 years). We estimate carbon uptake of construction waste by adding carbon uptake by construction waste concrete and construction waste mortar (see method).

3.4 Carbon uptake by cement kiln dust

Previous studies have shown that about 80% (52% to 90%) of the cement kiln dust removed from cement-producing kilns is diverted to landfills and 20% is beneficially re-used[14,60] *(Supplementary Data 18)*. Given the very small particle size, substantial carbonation occurs within the first 2 days of reaction in a landfill and complete carbonation is achieved within one year[16,17]. We estimate carbon uptake by CKD in different regions of the world based on the cement production, CKD generation rate, proportion of CKD treatment in landfill *(Supplementary Data4),* CaO proportion in CKD[14], and the fraction of CaO within fully carbonated CKD that has been converted to $CaCO_3$ (see method).

4 Annual and cumulative carbon uptake of cement materials

4.1 Annual carbon uptake of different cement materials

<u>Annual carbon uptake of cement concrete in different regions</u>

The annual carbon uptake of cement concrete in historic years is larger than that in recent years because carbon uptake of concrete occurs over many years and life cycle stages. We estimate that the annual carbon uptake of cement concrete materials in China, Europe, the U.S., and the rest of world have increasing from 0.002 million ton to 27.57million ton, 0.14 million ton to 9.61 million ton, 0.10 million ton to 4.08 million ton, and 0.01 million ton to 27.10 million ton from 1930 to 2013, respectively (*Supplementary Data 19*). Cumulative carbon uptake of by cement in concretes worldwide has increased from 0.25 million ton to 68.35 million ton.

<u>Annual carbon uptake of cement mortar in different regions</u>

The annual carbon uptake of cement mortar in recent years is greater than in historic years because the cement mortar is used in thin layers. The annual carbon uptake of cement mortar materials in China has increased from 0.01 million tons in 1930 to 90.95 million tons in 2013. The annual carbon uptake of cement mortar materials in Europe has meanwhile increased from 0.74 million ton in 1945 to 14.69 million tons in 1980, increasing little between 1980 and 1989, and then decreasing somewhat to between 14.62 million tons and 12.13 million tonsfrom1989 to 2013. The annual carbon uptake of cement mortar materials in the U.S. increased from 0.18 million tons in 1933 to a peak of 1.76 million tons in 2006, and then decreased to 1.43 million tons in 2013 (*Supplementary Data 19*). The annual carbon uptake of cement mortar materials in the rest of the world has increased from 1.70 million tons in 1930 to 148.96 million tons in 2013.

Annual carbon uptake of construction cement waste in different regions

The annual carbon uptake of cement mortar in recent years is larger than in historic years because most of the construction cement waste are small particles and finish carbonation in short time periods. The annual carbon uptake of construction cement waste materials in China, Europe, The U.S., and rest of world have increased from 479.37 tons to 5.87 million tons, 0.08 million tons to 1.02 million tons, 56.03 thousand tons to 0.19 million tons, and 4.73 thousand tons to 3.01 million tons between 1930 and 2013 (*Supplementary Data 19*). Over the same period, annual carbon uptake by cement in construction wastes worldwide has increased from 0.15 million tons to 9.85 million tons.

Annual carbon uptake of cement kiln dust in different regions

The annual carbon uptake of cement kiln dust in China has increasing from 1817.64 tons to 10.85 million tons between 1930 and 2013. The annual carbon uptake of cement kiln dust in Europe was 187.63 thousand tons in 1930, 1.82 million tons in 1980, and 1.38 million tons in 2013. The annual carbon uptake of cement kiln dust in the U.S. was 125.15 thousand tons in 1930 and increased to peak 0.45 million tons in 2005, before decreasing to 0.35 million tons in 2013. The annual carbon uptake of cement kiln dust in rest of world was 11.04 thousand tons in 1930, and increased to 5.56 million tons in 2011 before decreasing to 5.42 million tons in 2013 (*Supplementary Data 19*). The annual carbon uptake of cement kiln dust worldwide has increased from 0.32 million tons to 18.00 million tons between 1930 and 2013.

4.2 Annual carbon uptake of different cement material types in different regions

On an annual basis, carbon uptake by mortars is larger than that of concrete; uptake by concrete is greater than that of cement kiln dust; and cement kiln dust greater than that of construction wastes. The annual carbon uptake of cement materials in China has increased from 18.13 thousand tons in 1930 to 135.23 million tons in 2013. The annual carbon uptake of cement materials in Europe has increased from 1.72 million tons in 1930 to a peak of 24.20 million tons in 1989, before decreasing from 24.00 million tons in 1990 to 18.64 million tons in 1996, then back up to24.66 million tons in 2008, and finally down to 23.89 million tons in 2013. The annual carbon uptake of cement materials in the U.S. was increasing from 0.58 million tons in 1930 to the peak 6.30 million tons in 2006 with some fluctuations, decreasing to 5.03 million tons in 2009, and increasing to 6.05 million tons in 2013. The annual carbon uptake of cement materials in the rest of the world has increased from 0.10 million tons in 1930 to 79.99 million tons in 2013. Cumulative carbon uptake by cement materials in worldwide has increased from 2.42 million tons in 1930 to 245.15 million tons in 2013, dominated by carbon uptake of mortar. On average, more than 60% of carbon uptake was related to cement in mortar *(Supplementary Data 20)*. The majority of sequestration occurred in Europe and the U.S. prior to 1982, but cement materials used in China have absorbed more CO_2 than the other regions since 1994*(Supplementary Data 21)*. The annual carbon uptake of global cement material from 1930 to 2013 is shown in

Supplementary Data 22. There is legacy effect of accumulating cement stocks; on average, between 2000 and 2013, 19.2% of the carbon sequestered each year was absorbed by cement materials produced more than 5 years earlier and 13.8% produced more than 10 years earlier *(Supplementary Figure 4)*.

4.3 Cumulative process-based carbon emissions and carbon uptake of cement

Cumulative process CO_2emissions from world cement production are 10.4 GtC from 1930 to 2013 *(Supplementary Data 23)*, of which 6.6% was emitted in the U.S., 32.9% in China, 25.2% in Europe, and 35.4% in the rest of the world *(Supplementary Data 24)*. Our mean estimate of cumulative carbon uptake by cement materials worldwide between 1930 and 2013 is 4.50GtC, 1.40 GtC of which occurred in China, 1.18 GtC in Europe, 0.25 GtC in the U.S., and 1.67 GtC in the rest of the world. From cement material types, 1.24 GtC was absorbed by concrete, 2.73 GtC by mortars, 0.34 GtC by cement kiln dust, and 0.19 GtC by construction cement wastes *(Supplementary Table 11)*.

5 Uncertainty analysis

We use a Monte Carlo method as recommended by the 2006 IPCC guidelines for National Greenhouse Gas Inventories to evaluate uncertainty of CO_2 removal due to cement material carbonation [61]. We identify 26 causes of uncertainties associated with carbon sequestration estimates *(Supplementary Table 12)* which we vary across wide ranges to estimate the implications for carbon uptake. Each of the parameters and their ranges are discussed below.

1) Cement production vs consumption rate. We assume cement consumption is similar to production because the shelf life of cement is only 3 to 6 months, after which time unused cement will become waste. On average, cement production is less than 4.5% greater than consumption worldwide. Previous statistics suggest there is some regional variation, e.g., the difference in China averages 3.4% (from 0.1% to 10.8%) from 1996 to 2005 [62], the U.S. averages 4.5% (from -30.0% to 30.6%) from 1930 to 2013[11], Europe's average difference is 4.0% (from -15.1% to 12.8%) from 2003 to 2012[12]. For our uncertainty analysis, we assess differences between production and consumption as a normal distribution with mean value of 0.0%, a standard deviation of 4.0%, a maximum value of 30.6%, and a minimum value of -30.0% *(Supplementary Data 3)*.

2) Clinker to cement ratio. The 1996 IPCC Guidelines for National Greenhouse Gas Inventories suggested that the mean percentage of clinker in cement was 97%[63], and the 2006 IPCC Guidelines updated this to 75%[40]. Here, we vary the clinker to cement ratio from 75% to 97% in Weibull distribution with shape and scale parameters are 91.0% and 25, respectively.

3) CaO content in clinker. According to the 2006 IPCC Guidelines, the average CaO content in clinker is 65.0% (60.0- 67.0%)[40]. We therefore vary the CaO content in clinker assuming a triangular distribution with a mode value of 65.0%, a maximum value of 67.0%, and a minimum value of 60.0%.

4) MgO content in clinker. Previous studies have found that the average MgO content in clinker is approximately 2.5% (0-5.0%) because 5.0% addition of MgO will have little effect on the quaternary system[64,65]. We therefore vary the MgO content in clinker assuming a triangular distribution with a mode value of 2.5%, a maximum value of 5.0%, and a minimum value of 0.

5) Proportion of CaO converted to $CaCO_3$. We vary the proportion of CaO converted to $CaCO_3$ between 50.0%[9,18,42,43] and 90.0%[42], assuming a Weibull distribution with shape and scale parameters of 86.0% and 25, respectively, for concrete carbonation, and between 50.0%and 100% and a Weibull distribution with shape and scale parameters of 92.0% and 20, respectively, for mortar carbonation (the range of proportions derived from experimental tests; *Supplementary Data 16)*.

6) Concrete strength class distribution. *Supplementary Table 2* gives the range of Weibull distributions of concrete strength classes by region and *Supplementary Data 6* gives the relevant shape and scale parameters.

7) Proportion of cement for concrete. The proportions of cement used for concrete by region are assumed to be in Weibull distribution, with shape and scale parameters of 73.4% and 13, respectively, with a maximum value of 87.4%, and a minimum value of 47.2% for China *(Supplementary Data2)*; shape and scale parameters of 89.1% and 25.5, respectively, with a maximum value of 90.8% and a minimum value of 70.0% in the U.S.[11]; and shape and scale parameters of 74.9% and 14.8, respectively, and a maximum value of 87.8% and a minimum value of 62.3% in Europe[12,13] and the rest of the world.

8) Cement content of concrete. The cement content for concrete varies substantially by concrete strength class and use[12,18,20,22,66] *(Supplementary Data7)*. We vary the cement content of concrete in uniform distribution for each strength class with maximum and minimum values of 288 and 165 kg/m^3, respectively, for concrete strength classes of less than 15 MPa, 390 and 240 kg/m^3 for strength classes between 16 MPa and 23 MPa, 400 and 280 kg/m^3 for strength classes between 24 MPa and 35 MPa, and 670 and 300 kg/m^3 for strength classes greater than 35 MPa.

9) Carbonation rate coefficients for plain concrete. The carbonation rate coefficients for plain concrete by region are shown in *Supplementary Table 6*. We adopt maximum and minimum values from a range of studies of concrete carbonation rate coefficients[7,18,42,67] and from our field survey data. For concrete in indoor, outdoor exposed, and outdoor sheltered conditions, we use a maximum value of 15.0 $mm/\sqrt{\text{year}}$ and minimum value of 5.0 $mm/\sqrt{\text{year}}$ for concrete strength classes less than 15 MPa; maximum and minimum values of 9.0 and 2.5 $mm/\sqrt{\text{year}}$[5], respectively, for concrete strengths between 16 MPa and 22 MPa; maximum and minimum values of 6.0 and 1.5 $mm/\sqrt{\text{year}}$ for concrete strengths between 23 MPa and 35 MPa; and maximum and minimum values of 3.5 and 1.0 $mm/\sqrt{\text{year}}$ for concrete strength greater than 35 MPa. For concrete in buried and wet conditions, we assume a maximum and minimum values of 5.0 and 1.9 $mm/\sqrt{\text{year}}$ for concrete strength less than 15 MPa; values of 2.5 and 1.0 $mm/\sqrt{\text{year}}$ for concrete strength between 16 MPa and 22 MPa; values of 1.5 and 0.7 $mm/\sqrt{\text{year}}$ for concrete strength between 23 MPa and 35 MPa; and values of 1.0 and 0.3 $mm/\sqrt{\text{year}}$ for concrete strength larger than 35 MPa. In each case, we vary the coefficients assuming a uniform distribution.

10) The service life of buildings. We assume the range of building service life durations are in a Weibull distributed after a study by Kapur et al[38]. The shape and scale parameters are 42 years and 4, respectively, with maximum and minimum values of 73 and 4 years for China *(Supplementary Data9)*; shape and scale parameters are 75 years and 8, respectively, with maximum and minimum values of 90 and 50 years in Europe[24,68]; shape and scale parameters are 74.1 years and 4.4, respectively, with maximum and minimum values of 82.4 and 56.9 years in the U.S.[38]; and shape and scale parameters are 50 years and 3, respectively, with maximum and minimum values of 90 and 10 years in the rest of world[39].

11) Distribution of waste concrete particle size. Based on 179 field survey datasets from China, we assume the particle size of waste concrete is uniformly distributed. The ranges of particle sizes by region are taken from the literature and are given in *Supplementary Data 12*[18,39,46,48].

12) Waste concrete exposure time during demolition stage. Based on 985 field survey demolition projects in China, the waste concrete exposure time during demolition stage is assumed to be distributed according to a Weibull distribution *(Supplementary Data 10)*. The shape and scale parameters for China are 0.5 years and 4, respectively, with maximum and minimum values of 0.8 and 0.1 years. Based on previous studies, for Europe, the U.S. and the rest of the world, we assume similar shape and scale parameters (0.5 years and 4, respectively), with maximum and minimum values of 1.0 and 0.1 years[18,24,34].

13) Correction factors related to cement additives. Cement additives may increase the carbonation rate of concrete and mortar[18,35]. Correction factors related to such additives are estimated in Weibull distribution with globally applicable shape and scale parameters of 1.16 and 20, respectively, and maximum and minimum values of 1.3 and 1.0, respectively *(Supplementary Table 4)*[18].

14) Correction factors for CO_2 concentration. Elevated CO_2 concentrations in the ambient atmosphere of industrial areas[69] and areas near roads will also increase the carbonation rate[25,26]. We assume correction factors for these elevated CO_2 concentrations are Weibull distributed with globally applicable shape and scale parameter of 1.18 and 25, respectively, and maximum and minimum values of 1.41 and 0.93, respectively *(Supplementary Table 3)*.

15) Correction factors for cover and coating. Application of surface coating on concrete can reduce carbonation rates[27]. If the concrete strength and age are known, some studies have found that surface coating such as paint lowers the rate of carbonation by 0-50%[9,18,19,42]. We assume the correction factors for such coverings and coatings are Weibull distributed with globally applicable shape and scale parameters of 1.0 and 6.0, respectively, and maximum and minimum values of 1.0 and 0.5, respectively *(Supplementary Table 5)*.

16) Proportion of cement used for mortar. The proportion of cement for mortar is estimated in Weibull distribution. The shape parameter and scale parameter are 30.8% and 12, respectively, with maximum value is 91.7% and minimum value is 10.0% in China[62]. The shape parameter and scale parameter are 29.0% and 12 respectively with maximum value is 37.1% and minimum value is 12.0% in Europe[12]. The shape parameter and scale parameter are 13.2% and 12.5 respectively with maximum value is 29.6% and minimum value is 9.1% in the U.S.[11]. The Weibull distribution parameters of proportion of cement for mortar in rest of world refer to situations of Europe *(Supplementary Data 2)*.

17) Proportion of mortar utilization types. Mortar utilization is mainly in three types: (1) rendering, plastering and decorating, (2) masonry, and (3) maintenance and repairing[50]. Most mortar cement is used for rendering, plastering and decorating[51]. The proportion of mortar utilization types are estimated in Weibull distribution for each region, as shown in *Supplementary Data 14*.

18) Thickness of different mortar utilizations. The cement mortar is used in thin layer with large exposure area[50]. The thickness of mortar utilizations showed obvious impacts on carbon sequestration. The mortar thickness for rendering, plastering and decorating is estimated in Weibull distribution with shape parameter and scale parameter are 22 mm and 4, respectively, and maximum value is 50 mm and minimum value is 3 mm. The mortar thickness for masonry is estimated in Weibull distribution with shape parameter and scale parameter are 11 mm and 8, respectively, and maximum value is 20 mm and minimum value is 5 mm. The mortar thickness for maintenance and repairing is estimated in Weibull distribution with shape parameter and scale parameter are 26.8 mm and 7, respectively, and maximum value is 50 mm and minimum value is 10 mm[51] *(Supplementary Table 10)*.

19) Proportions of masonry wall with rendering mortar. The proportions of walls with both sides rendered, one side rendered, and no rendering are derived from 1144 survey projects in China. Based on the survey results, we vary the proportions of masonry wall with various extents of rendering as a triangular distribution with mode value of 60% (maximum is 90% and minimum is 40%) for wall with both sides rendered, mode value of 30% (maximum is 50% and minimum is 10%) for wall with one side rendered, and mode value of 10% (maximum is 20% and minimum is 0%) for walls without rendering. We assume these ranges apply globally *(Supplementary Data 17)*.

20) Wall thickness. Wall thicknesses worldwide range from 60 to 610 mm[9,22,57], with most between 100 and 490 mm. Wall thicknesses greater than 500 mm are used for defense facilities, river & harbor development & control, and dams and reservoirs[9]. For this study, we assume uniformly distributed thickness between610 and 60 mm *(Supplementary Data 11).*

21) Carbonation rate coefficients for mortar. Based on 1600 field experiments we conducted in China, we assume worldwide mortar carbonation rate coefficients in triangular distribution with a mode value of 19.6 $mm/\sqrt{\text{year}}$, maximum value of 36.8 $mm/\sqrt{\text{year}}$, and minimum value of 6.1 $mm/\sqrt{\text{year}}$ *(Supplementary Data 15)*.

22) Proportion of cement loss in construction stage. Construction budget standards[22] and survey data[58] indicate that between 1 and 3% of cement is lost during construction. We therefore vary the percent lost assuming a triangular distribution spanning this range and with a mode value of 1.5% *(Supplementary Data 4)*.

23) Construction waste concrete carbonation time. Most construction waste concrete is in the form of small particles that are either recycled as backfill or landfilled after the completion of building projects[37,59]. The construction waste concrete carbonation time is estimated in triangular distribution with mode value is 5 years, maximum value is 10 years, and minimum value is 1 year.

24) CKD generation rate based on clinker. Cement kiln dust (CKD) production is estimated as a fraction of clinker production[14], which fraction is varied in a triangular distribution with a mode value of 6.0%, a maximum value of 11.5% (representative of wet kilns), and a minimum value of 4.1% (representative of preheater/precalciner kilns)[14].

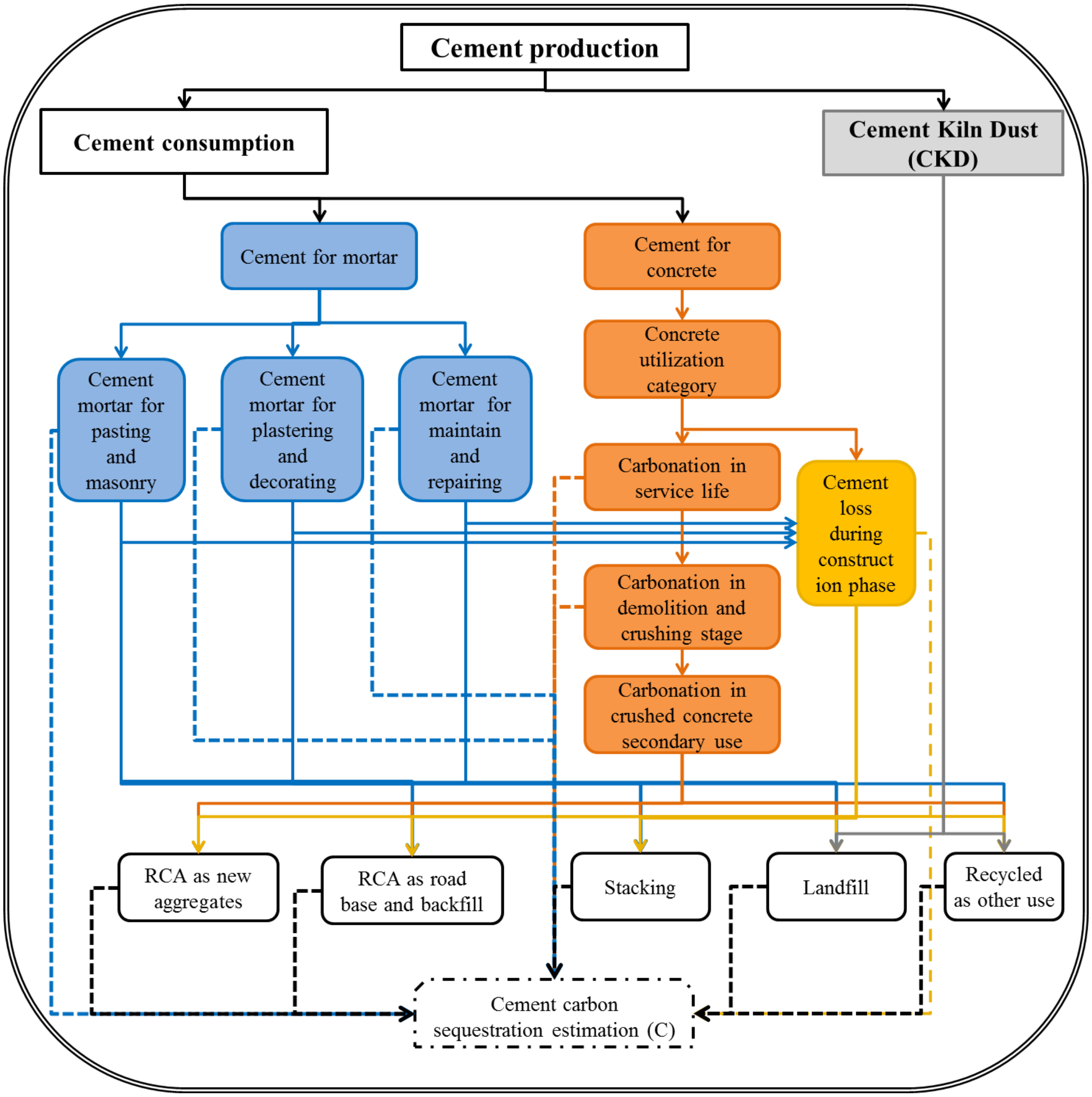
25) Proportion of CKD sent to landfill. Previous studies have shown that about 80% of cement kiln dust is diverted to landfills after removal from the kilns, and 20% is beneficially re-used[15,60]. We vary this landfill proportion assuming a triangular distribution with mode value of 80.0%, maximum value of 90.0%, and minimum value of 52.0%[14].

26) CaO content in CKD. The average CaO content in CKD is about 44.0%[14,70]. We vary this content assuming CaO content of CKD is normally distributed with a mean value of 44.0%, a standard deviation of 8.01, a COV of 18%, a maximum value of61.2%, and a minimum value of 19.4%[60,70].

Based on the uncertainty analysis described above, we find the mean value of carbon uptake from cement materials in China is 0.14 Gt C with a 2σ standard deviation of 15.4% in 2013. The mean value of carbon uptake from cement materials in the United States is 0.01 Gt C with a 2σ standard deviation of 15.6% in 2013. The mean value of carbon uptake from cement materials in Europe is 0.02 Gt C with a 2σ standard deviation of 13.7% in 2013. The mean value of carbon uptake from cement materials in the rest of world is 0.09 Gt C with a 2σ standard deviation of

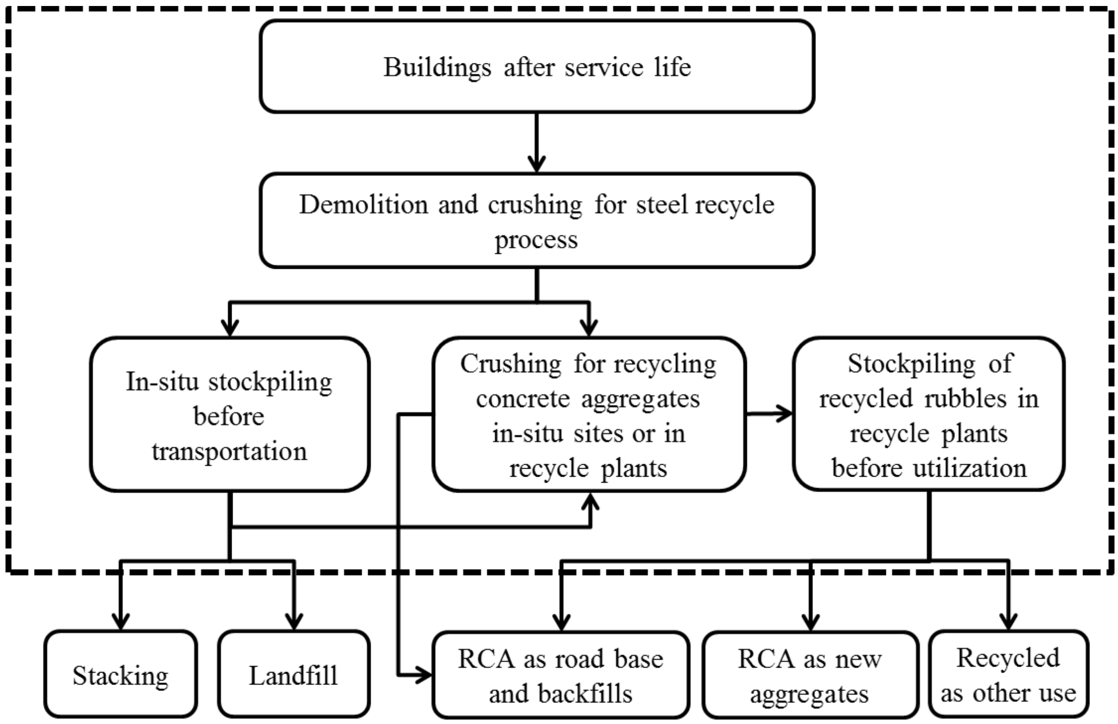
15.1% in 2013. Worldwide, the mean value carbon uptake from cement materials is 0.25 Gt C (2σ standard deviation of 10.0%) in 2013.

Supplementary Figure 1 **The framework and system boundary of carbonating cement materials.**



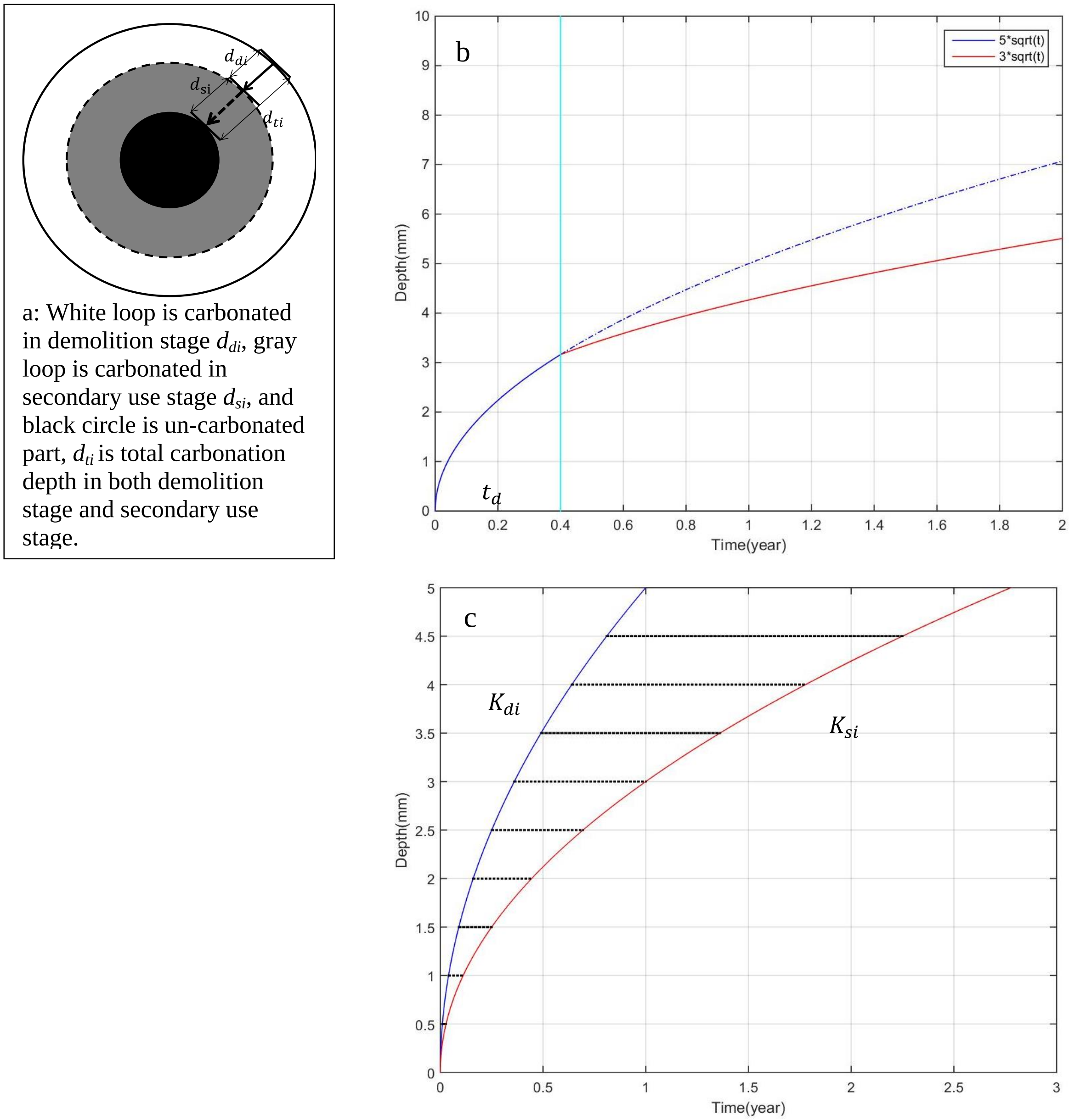
*The solid line presents material flow, the dot line presents carbon sequestration flow, and the double solid line presents the system boundary of the study.

Supplementary Figure 2 **The processes of cement material flow in demolition stage***



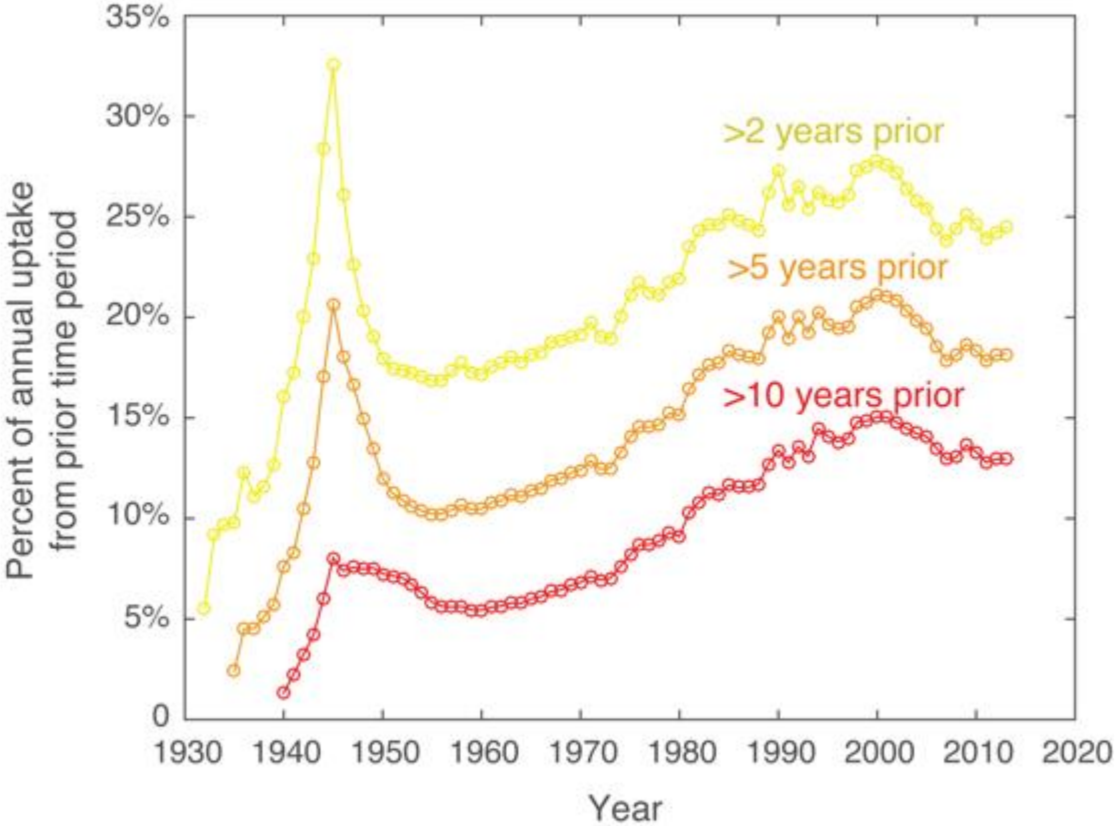
* The content within dotted framework is the process of demolition stage and the content out of the dotted framework is the fate of waste concrete in secondary use stage.

Supplementary Figure3 **Carbonation model in secondary use stage and carbonation time delay for same carbonation depth under air exposure condition and buried condition**



a is a sketch of the continued carbonation depth in treatment and secondary use stage; **b** shows the carbonation depth of C15 concrete rubble in the demolition stage and secondary use stage (the blue line is the carbonation rate in open air and the red line is the carbonation rate in buried condition), t_d is average exposure time during demolition stage; **c** shows the time lag (Δt_i) for same carbonation depth under air exposure condition and buried condition for C15 concrete rubble. k_{di} is the carbonation rate coefficient in air exposure condition and k_{si} is the carbonation rate coefficient in buried exposure condition.

Supplementary Figure 4 **The time lag effect of annual uptake from prior time period**



Supplementary Table 1 **Categories of cement use by region**

Countries and regions	**Cement for concrete (%)**			**Cement for mortar (%)**			**Cement loss in construction (%)**			**Data sources** (Reference)
	Ave	Max	Min	Ave	Max	Min	Ave	Max	Min	
China	69.7	88.5	6.8	28.8	91.7	10.0	1.5	3.0	1.0	SI data 2,
Europe	71.1	86.5	61.4	27.4	29.6	9.1	1.5	3.0	1.0	[12,13,18], SI data2
United States	86.0	89.4	68.9	12.5	37.1	12.0	1.5	3.0	1.0	[11], SI data 2
Rest of world	71.1	86.5	61.4	27.4	37.1	9.1	1.5	3.0	1.0	Refer to Europe

Supplementary Table 2 **Categroies of concrete strength class use by region**

Countries and regions	**Concrete strength class percent (%)**				**Data sources** (Reference)
	⩽C15 (range)	**C16- C22 (range)**	**C23- C35 (range)**	**>C 35 (range)**	
China	14.9 (0-33.5)	12.5 (0-25.8)	66.2 (41.6-82.8)	10.4 (0-23.4)	SI data6
Europe	5.3 (2.9-8.0)	39.0 (18.9-54.0)	45.3 (32.0-62.9)	10.4 (8.0-13.5)	[12,13,18]
U.S.	21.2 (0-40.0)	38.8 (5.0-60.0)	27.7 (20.0-80.0)	12.3 (10.0-15.0)	[12,20,21]
Rest of world	5.3 (2.9-8.0)	39.0 (18.9-54.0)	45.3 (32.0-62.9)	10.4 (8.0-13.5)	Refer to Europe

Supplementary Table 3 **CO_2 concentration and correction value under different environment* (β_co_2)**

Location	CO_2 concentration (ppm)	modified parameter*
Urban	625	1.20
Rural	300	1.00
Seaside	225	0.93
Industrial area	1200	1.41
Road	1200	1.41
Buried	3000	1.00

*The carbonation depth is in proportion to $\sqrt{CO_2\ concentration}$ according to Papadakis *et al.* *and Yoon et al.* [25,26]

Supplementary Table 4 **Correction factors for different powder additions to be multiplied by the carbonation rate coefficients provided for concrete (β_ad)**

Type of addition	**Amount of addition (wt.%)***						Data sources (Reference)
	0-10%	10-20%	20-30%	30-40%	40-50%	60-80%	
Limestone		1.05	1.1				18
Fly-ash		1.05		1.1			
Silica-fume	1.05	1.1					
GGBF-Slag	1.05	1.1	1.15	1.2	1.25	1.3	

*wt.% is the weight percentage of addition in cement.

Supplementary Table 5 **Coating and cover and correction values under different environment (β_cc).**

Coating and cover type	**correction values**	**Data sources** (Reference)
no coating and cover	1	19,24,28,31-33,42
indoor concrete coating	0.7	
outdoor concrete coating	0.5	
Infrastructure concrete if painted	1	

Supplementary Table 6 **Carbonation rate coefficients for various concrete strength classes and concrete exposure conditions**

Region	**Exposure condition**	**Compressive strength (mm/(year)$^{0.5}$)**				**Data source** (Reference)
		⩽15 MPa	**16-22 Mpa**	**23-35 MPa**	**>35MPa**	
Europe (plain concrete)	**Exposed**	5	2.5	1.5	1	[18,42]
	Sheltered	10	6	4	2.5	
	Indoors	15	9	6	3.5	
	Wet	2	1	0.75	0.5	
	Buried	3	1.5	1	0.75	
China (plain concrete)	**Exposed**	6.1	3.9	2.4	1.3	SI data 8
	Sheltered	9.9	7.1	4.8	2.5	
	Indoors	13.9	9.8	7.0	4	
	Wet	3.8	1.9	1.0	0.5	
	Buried	1.9	1.0	0.7	0.3	
U.S.	**Uncoated**	7.1	6.9	3.8-5.4	2.5	[9]
	Coated	N/A	3.5	1.9-2.7	N/A	

Supplementary Table 7 **The average service life, demolition stage, and secondary use stage (years) in different countries.**

Countries and regions	**Average Service life t_l (range)**	**Average Demolition stage t_d (range)***	**Average secondary use stage t_s (range)**	**Assessment time *t*****	**Data sources** (Reference)
China	35 (4-73)	0.4 (0.1-0.8)	64.6	100	[36,37], SI data 9 and SI data 10
U.S.	65 (56-73)	0.4 (0.1-0.8)	34.6	100	[38]
Europe	70 (50-90)	0.4 (0.1-0.8)	29.6	100	[18,24,34,42,68]
Rest of world	40 (10-90)	0.4 (0.1-1.0)	59.6	100	[39,46,71]

* The $\boldsymbol{t_d}$ in China is estimated based on the Chinese studies[36,37] and field survey data (Supplementary Data 9 and Supplementary data 10).

** $t = t_l + t_d + t_s$

Supplementary Table 8 **The waste concrete secondary use methods and proportions**

<table>
<tr><th>Secondary use methods of demolished concrete</th><th>China Average(range) %</th><th>Europe Average (range) %</th><th>U.S. Average (range) %</th><th>Rest of world Average (range)%</th></tr>
<tr><td>RCA for new concrete</td><td>0.01 (0-1)</td><td>0.72 (0.3-1.8)</td><td>3.60 (2.5-5)</td><td>1.00 (0-2)</td></tr>
<tr><td>RCA for road base, backfills materials and others use</td><td>2.30 (2-5)</td><td>60.42 (40-80)</td><td>51.00 (40-60)</td><td>24.0 (14-34)</td></tr>
<tr><td>Landfill</td><td rowspan="2">97.69 (80-98)</td><td>38.86 (20-70)</td><td>40.00 (30-50)</td><td rowspan="2">75.00 (60-85)</td></tr>
<tr><td>Dumped and Stacking</td><td>0</td><td>0</td></tr>
<tr><td>Asphalt concrete</td><td>0 (0-1)</td><td>0 (0-1)</td><td>5.40 (4-6.5)</td><td>0 (0-1)</td></tr>
<tr><td>Data sources
(Reference)</td><td>[44]</td><td>[48]</td><td>[45]</td><td>[39,46]</td></tr>
</table>

RCA: Recycling Concrete Aggregate

Supplementary Table 9 **The waste concrete treatment methods and particle size proportion in different regions**

Demolished and crushed concrete treatment types	**Particle size grading**	**Particle size distribution percentage in different regions (%)**			
		China (range)*	Europe (range)**	The U.S. (range)***	Rest of world (range) ****
RCA for new concrete	<5mm	14.9(2.1-20.0)	29.4(22.5-36.0)	29.4(10.0-36.0)	24.1(15.0-37.0)
	5-10mm	25.1(17.5-41.2)	13.8(12.5-15.0)	13.8(5.0-30.0)	17.0(12.0-23.0)
	10-20mm	40.6(32.0-45.0)	39.2(20.0-44.0)	39.2(20.0-44.0)	33.9(24.0-46.0)
	20-40mm	19.4(10.0-26.7)	17.6(5.0-45.0)	17.6(10.0-30.0)	25.0(16.0-39.0)
RCA for Road base and others	<1mm	11.7(5.1-20.0)	15.7(10.0-21.0)	15.7(10.0-21.0)	16.1(10.0-24.7)
	1-10mm	26.9(20.0-36.7)	27.5(25.0-30.0)	27.5(25.0-30.0)	25.0(20.3-28.0)
	10-30mm	42.0(35.6-60.0)	39.2(20.0-44.0)	39.2(20.0-44.0)	42.3(35.3-51.3)
	30-53mm	19.4(0-28.0)	17.6(5.0-45.0)	17.6(5.0-45.0)	16.7(10.7-26.0)
Landfill and Stacking	<10mm	17.8(12.2-25.6)	17.8(12.2-25.6)	17.8(12.2-25.6)	17.8(12.2-25.6)
	10-30mm	27.1(19.5-35.4)	27.1(19.5-35.4)	27.1(19.5-35.4)	27.1(19.5-35.4)
	30-50mm	17.3(10.6-22.5)	17.3(10.6-22.5)	17.3(10.6-22.5)	17.3(10.6-22.5)
	>50mm	37.8(24.8-48.4)	37.8(24.8-48.4)	37.8(24.8-48.4)	37.8(24.8-48.4)
RCA for bituminous concrete	<5mm	14.9(2.1-20.0)	29.4(22.5-36.0)	29.4(10.0-36.0)	24.1(15.0-37.0)
	5-10mm	25.1(17.5-41.2)	13.8(12.5-15.0)	13.8(5.0-30.0)	17.0(12.0-23.0)
	10-20mm	40.6(32.0-45.0)	39.2(20.0-44.0)	39.2(20.0-44.0)	33.9(24.0-46.0)
	20-40mm	19.4(10.0-26.7)	17.6(5.0-45.0)	17.6(10.0-30.0)	25.0(16.0-39.0)
Data sources (Reference)		SI data12	[13,18,48]	[38,45]	[39,46]

* The crushed concrete particle size distribution in China is based on field survey data from 35 cities in China. The max diameter of recycled concrete aggregates for new concrete is 32.5mm in China and 40mm in Europe, U.S., and other countries.

** We use the waste concrete treatment proportion, particle size, and proportion in Nordic countries for European countries' situation.

*** The particle size and proportion for RCA for new cement concrete and asphalt concrete and RCA for road base materials and backfill refer to European situation.

**** The crushed concrete particle size distribution situations in other countries are estimated based on Japan and Korea.

Supplementary Table 10 **The thickness of different cement mortar utilization types**

Cement mortar utilization types	Cement mortar sub-utilization types	Thickness range of utilization d_T (mm)	Median thickness of utilization (mm)	Data sources (Reference)
Rendering, plastering, decorating and finishing	Rendering	10-30	20	50,51
	Plastering	1-5	3	50
	Tile adhesive	15-30	20	
	Tile grout	3-30	15	
	The exterior thermal insulation	5-10	8	
	power paints and waterproofing	1-2	1	
	self-leveling underlayment	5-30	20	
	screeds	30-80	50	
Masonry	Masonry	5-15	10	
Maintenance and repairing	Maintenance and repairing	10-30	25	

Supplementary Table 11 **The total accumulated carbon uptake from cement materials from 1930 to 2013 in the world**

Countries and regions	**Carbon uptake from cement materials (Gt carbon)**	**Carbon uptake from concrete (Gt carbon)**	**Carbon uptake from mortar (Gt carbon)**	**Carbon uptake from construction waste (Gt carbon)**	**Carbon uptake from CKD (Gt carbon)**
China	1.40	0.28	0.94	0.06	0.11
Europe	1.18	0.33	0.71	0.05	0.09
U.S.	0.25	0.12	0.09	0.01	0.02
The rest of world	1.67	0.50	0.98	0.07	0.12
Total world	4.50	1.24	2.73	0.19	0.34

Supplementary Table 12 **The causes and parameters for uncertainty analysis**

Types of cement utilization	Types of uncertainty	Causes of uncertainty	Distribution pattern	Parameter Description and value range				Data sources (Reference)
Cement	Activity data	1. Cement production vs consumption deviation rate	Normal	mean	Standard deviation	max	min	[11,12,62] SI data 3
				0	4.0%	30.6%	-30.0%	
	Impact factors of carbonation	2.Clinker to cement rate	Weibull	a	b	max	min	[40,63]
				91.0%	25	97.0%	75.0%	
	Impact factors of carbonation	3.CaO Content in Clinker	Triangular	mode		max	min	[40]
				65.0%		67.0%	60.0%	
	Impact factors of carbonation	4. MgO Content in Clinker	Triangular	mode		max	min	[64,65]
				2.5%		5.0%	0	
	Impact factors of carbonation	5. Proportion of CaO converted to $CaCO_3$	Weibull	a	b	max	min	
		for concrete		86.0%	25	90.0%	50.0%	[9,18,41-43]
		for mortar		92.0%	20	100%	50.0%	SI data16
Concrete	Activity data	6. Concrete strength class distribution	Weibull	a	b	max	min	[12,18]
				See SI data 6				SI data 6
	Activity data	7. Proportion of Cement for concrete	Weibull	a	b	max	min	[11-13] SI data 2
		for China		73.4%	13	87.4%	47.2%	
		for Europe		74.9%	14.8	87.8%	62.3%	
		for U.S.		89.1%	25.5	90.8%	70.0%	
		for rest of world		74.9%	14.8	87.8%	62.3%	
	Activity data	8. The cement content for concrete (kg/m^3)	Uniform			max	min	[12,18,20,22,66]
						See SI data 7		SI data 7
	Activity data	9. Carbonation rate coefficients for plain concrete (mm/$\sqrt{year}$)	Uniform			max	min	[7,9,18,19,34,39,67]
						See SI data 8		SI data 8
	Activity data	10. Service life of building	Weibull	a	b	max	min	[24,38,39,68] SI data 9
		for China (year)		42	4	73	4	
		for Europe (year)		75	8	90	50	
		for U.S. (year)		74.1	4.4	90	45	
		for rest of world(year)		50	3	90	10	

	Activity data	11. Proportion of waste concrete particle distribution	Uniform			max	min	[18,39,46,48]
						See SI data 12		SI data 12
	Activity data	12. The waste concrete exposure time in demolition stage (year)	Weibull	a	b	max	min	[18,24,34]
				0.5	4	1	0.1	SI data 10
	Impact factors of carbonation	13. Correction factors of cement additions	Weibull	a	b	max	min	[18,35]
				1.16	20	1.3	1	SI data 5
	Impact factors of carbonation	14. Correction factors of CO_2 concentration	Weibull	a	b	max	min	[25,26]
				1.18	25	1.2	0.93	
	Impact factors of carbonation	15. Correction factors of cover and coating	Weibull	a	b	max	min	[9,18,19,42,72,73]
				1	6	1.0	0.5	
Mortar	Activity data	16. Proportion of cement for mortar	Weibull	a	b	max	min	[11-13] SI data 2
		for China		30.8%	12	53.8%	12.6%	
		for Europe		29.0%	12	37.7%	12.2%	
		for U.S.		13.2%	12.5	30.0%	9.2%	
		for rest of world		29.0%	12	37.7%	12.2%	
	Activity data	17. Proportion of mortar utilization types	Weibull	a	b	max	min	[50,51]
				See SI data 13				SI data 13
	Activity data	18. Thickness of different mortar utilizations	Weibull	a	b	max	min	[50,51] SI Table 10
		for rendering, plastering and decorating (mm)		22	4	80	3	
		for Masonry (mm)		11	8	20	5	
		for maintenance and repairing (mm)		26.8	7	50	10	
	Activity data	19. Proportions of masonry wall with render	Triangular	mode		max	min	SI data 17
		for both sides render		60%		90%	40%	
		for one side render		30%		50%	10%	
		for no render		10%		20%	0%	
	Activity data	20. Wall thickness (mm)	Uniform			max	min	[22,57]
						610	60	SI data 11
	Sequestration	21. Carbonation rate	Triangular	mode		max	min	[54]

	factor of mortar	coefficients for mortar (mm/$\sqrt{\text{year}}$)		19.6		36.8	6.1	SI data 15
Construction cement waste	Activity data	22. Proportion of cement loss in construction stage	Triangular	mode		max	min	[22,58]
				1.5%		3.0%	1.0%	
	Activity data	23. Construction waste concrete carbonation time (year)	Triangular	mode		max	min	[37,59]
				5		10	1	
Cement Kiln Dust (CKD)	Activity data	24. CKD generation rate based on clinker	Triangular	mode		max	min	[14]
				6.0%		11.5%	4.1%	
	Activity data	25. Proportion of CKD sent to landfill	Triangular	mode		max	min	[15,60]
				80.0%		90.0%	52.0%	
	Impact factors of carbonation	26. CaO content in CKD	Normal	Mean	Standard deviation	max	min	[60,70]
				44.0%	8.01	61.23%	19.40%	

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